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# Strong and weak seeded four-wave mixing in stretched gas-filled hollow capillary fibers

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## ABSTRACT

We report a remarkably efficient experimental scheme for the generation of high energy ultra-short pulses by means of four-wave mixing in long stretched hollow capillary fibers filled with helium. We thoroughly investigate the role of strong and weak seeding fields in a degenerate up-conversion scheme to the deep ultraviolet. In the weak seed regime we demonstrate the tunable emission of up to 30  $\mu\text{J}$  in ultrashort pulses ( $\sim 8$  fs) in the 250-300 nm range, corresponding to pump energy conversion of up to 30%, from pump pulses with energies readily available from high-average power lasers. In the strong seed regime, we obtain higher pump conversion efficiencies, up to 42%, together with a spectral bandwidth supporting few femtosecond pulses and a record high deep-ultraviolet pulse energy exceeding 70  $\mu\text{J}$ . The energy can be further scaled by using stretched hollow-core fibers with larger core diameters.

**Keywords:** Ultrafast phenomena, nonlinear fiber optics, ultraviolet wavelength conversion, four-photon interactions.

## 1. INTRODUCTION

Despite being an effect more than 50 years old <sup>1</sup>, four-wave mixing (FWM) processes have an unexplored potential. This is especially the case in stretched hollow-core fibers (HCF) <sup>2</sup>, which provide a fully energy-scalable platform for nonlinear optics <sup>3</sup>—for example, we recently demonstrated the energy scaling of soliton-driven nonlinear effects in stretched-HCF, such as pulse self-compression and tunable dispersive wave emission <sup>4</sup>. Pioneering works on FWM in rigid gas-filled hollow-core fibers <sup>5</sup>, have demonstrated up-conversion efficiencies up to 30% and a maximum energy of up to 65  $\mu\text{J}$  in a chirped-pulse configuration <sup>6</sup>. Here we focus on the nonlinear effects driven by the seeding regime in stretched-HCF in order to optimize the up-conversion efficiencies of un-chirped pulses.

## 2. EXPERIMENTS AND DISCUSSION

We investigate experimentally the role of the seeding field in degenerate four-wave mixing in a 1.35 m long, 150  $\mu\text{m}$  inner diameter, stretched-HCF filled with helium. The pump pulse is obtained by frequency doubling a commercial 1 kHz Ti:Sa CPA laser system operating at 800 nm in a 100  $\mu\text{m}$  BBO crystal. A small portion of the 800 nm laser energy is used as the seed pulse and both the seed and pump pulses are linearly co-polarized. The pump and seed pulses have temporal durations at the fiber input end of 35 fs and 45 fs, as retrieved from our home-built self-diffraction and second-harmonic FROG devices. The powers at the fiber input and output end have been carefully monitored and cross-checked with both calibrated power-meters and a spectrometer which was absolutely calibrated in both spectral power density and wavelength.

Fig. 1 offers an overview of the generated powers in the up-converted anti-stokes (AS) beam in the spectral band from 240 nm to 320 nm. The gas filling pressure, pump and seed energies all drastically alter the generation process and the up-conversion efficiencies. At low seed energy ( $P_s < 12 \mu\text{J}$ )—in what we will call the weak seed regime—the effect of the cross-phase modulation induced by the seed on the pump and AS beam is negligible. In this regime the maximum AS generation shifts towards lower and lower pressures as the pump energy is raised (see e.g. Fig. 1c). This is the result of compensating the pump-driven nonlinear phase shift with linear dispersion through the He gas pressure <sup>7</sup>. On the other hand, for higher seed energies (Fig. 1d-f) the optimal phase-matching pressure and therefore parametric gain ( $\propto$  slope in Fig. 1a-f) shifts towards larger He pressures for a given pump energy, with an overall optimal pressure of around 1.9 bar in the strong seed regime (purple line, Fig. 1d-f for  $P_s > 70 \mu\text{J}$ ) in contrast to 1.6 bar in the weak seed regime (green line, Fig. 1a-c). This is highlighted by a direct comparison in Fig. 2 (a,e).

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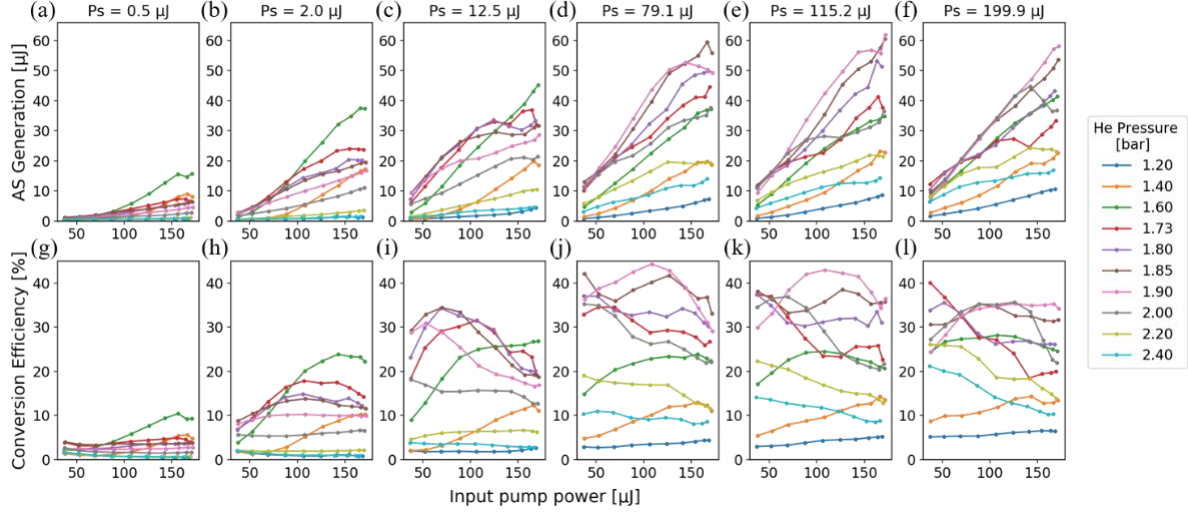


Figure 1. Overview of the generated power (AS, top row) and corresponding energy conversion efficiencies (lower row) in the anti-stokes band (240-320 nm) at the fiber output end as function of input pump powers. The different columns correspond to different seed powers ( $P_s$ ) at the fiber input end, while line colors indicate the different He pressures. The bottom row shows the corresponding AS energy conversion efficiencies calculated from the input pump powers. Every experimental point shown here is obtained for an optimal temporal delay (maximizing the AS power) between the seed and pump pulses.

It is important to remark how the use of long interaction lengths and the weak dispersion provided by gas-filled capillaries, make it possible to drive the FWM process even at moderate pump energies ( $\sim 60 \mu\text{J}$ ) with remarkably low seed energies ( $< 10 \mu\text{J}$ ). These relaxed energy requirements are well within current technology of high-average power lasers, such as thin-disk or fiber based laser systems, and so the large conversion efficiency demonstrated here (up to  $\sim 35\%$ ) provide a route for significant average-power scaling in the deep ultraviolet. Moreover, despite using fixed two-color input fields, the induced self-phase modulation of the pump and induced cross-phase modulation on the seed, make it possible to tune the AS generation across a spectral band covering the range from 250-300 nm, as shown in Fig. 2b, without the need of an external tunable seed but simply by adjusting the gas-filling pressure (Fig. 2b) or relative pump-seed delay (not shown here).

In the strong seed regime, depicted in Fig. 1d-f and Fig. 2e-g, we optimized the system for the maximal energy conversion and throughput in the AS band, by using a stronger seed pulse to carefully balance the pump induced cross-phase modulation, with the seed induced phase modulation, in order to optimize the pump depletion, which in this regime can reach up to 80% (not shown here). As shown with purple lines in Fig. 1d-f, corresponding to an He pressure of 1.9 bar, the parametric gain is now maximal for larger seed and pump powers, as clearly evident by the larger conversion efficiencies in Fig. 1j-k. The shift towards higher (lower) pressures in the strong (weak) seed regime is corroborated by a direct comparison in Fig. 2a and Fig. 2e which shows the AS power as a function of the pump power and He pressure for a weak seed of just  $1.9 \mu\text{J}$  and strong seed of  $113 \mu\text{J}$ . In order to find the overall optimal condition for AS power generation, we fine tune the seed power by keeping fixed the pump power, as shown in Fig. 2g. In this way we achieved an overall maximal AS generation of  $70 \mu\text{J}$  with 42% conversion efficiency from the input pump energy.

Fig. 2b and Fig. 2f show several snapshots of the generated AS spectra for different gas filling pressures in the weak and strong seed regime, obtained by maximizing the AS power via pump-seed delay and pump power. While in the weak seed regime the pulse bandwidth is nearly constant across the 250-300 nm band as result of the self and cross phase modulation driven by the pump; in the strong seed regime, instead, the spectral shape and bandwidth strongly depends on the seed power and pump-seed delay. The possibility of controlling the spectral shape, and even the spectral phase<sup>8</sup>, together with the large conversion efficiencies, marks a striking difference with alternative up-conversion schemes, such as dispersive wave emission and nonlinear crystals. From the experimental and retrieved FROG traces (0.6% error) from a home-build self-diffraction FROG we obtain a pulse duration for the AS pulse of 8 fs in the weak seed regime, close to the bandwidth limit of 7.7 fs. A shorter bandwidth-limited pulse duration can be achieved in the strong seed regime ( $< 5$  fs) and even shorter or much longer pulses are expected with more advanced seeding schemes. Control over the generated spectral bandwidth and tuning of the pulse duration are under study and our latest advancements will be presented.

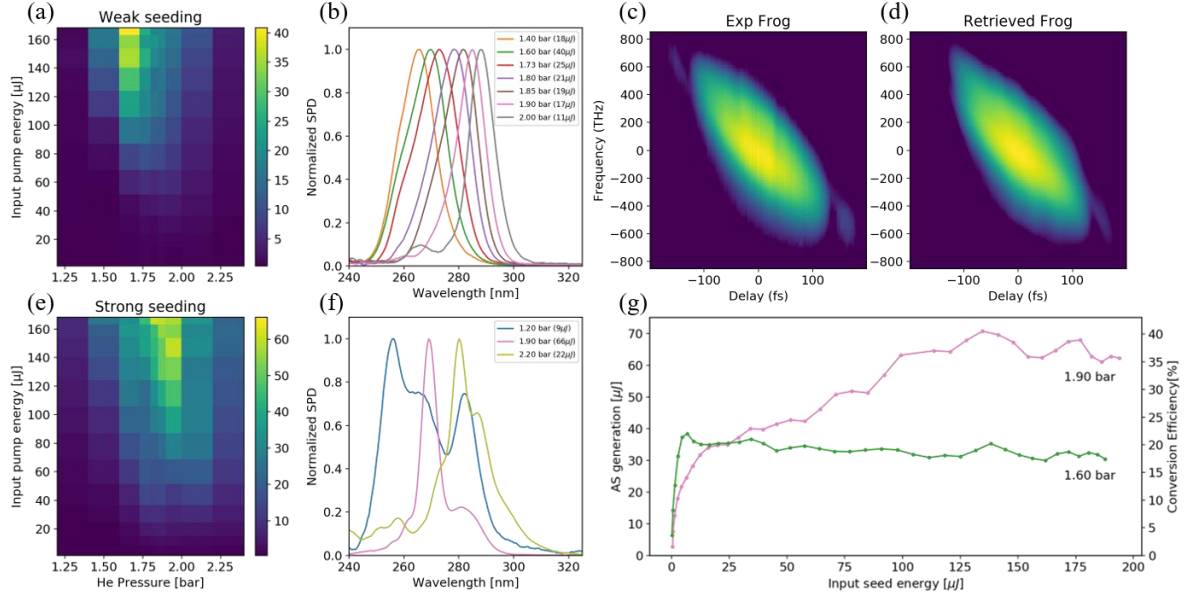


Figure 2. Pseudo color map of the experimentally generated AS powers in the weak (a) and strong seed regime (e) as function of the input pump energy and He pressure. Tuning curve in the weak (b) and strong (f) seed regime of the spectra at different gas pressure providing the maximal AS power as shown in the inset. (c) Measured and (d) retrieved SD-FROG traces of the AS pulse. (g) AS power as function of the input seed power at the maximal pump power investigated in this work (170  $\mu\text{J}$ ) and optimal pressures of 1.6 and 1.9 bar for the weak and strong seed case.

### 3. CONCLUSIONS

In this contribution we have investigated the effect of a strong and weak seeded four-wave mixing scheme in helium filled and stretched-HCF for up-conversion to the deep-ultraviolet band. We demonstrated the generation of ultrashort pulses ( $< 8$  fs) in the deep-ultraviolet with remarkably high-conversion efficiency, up to 30% in the regime of moderate pump energies ( $< 60$   $\mu\text{J}$ ) and low seed energies ( $< 10$   $\mu\text{J}$ ), and up to 42% by using stronger pump and seed pulses, reaching a record high deep ultraviolet pulse energy exceeding 70  $\mu\text{J}$ . Seeded four-wave mixing in gas-filled and stretched hollow-fiber represents a robust route for scaling up the average or peak power in spectral regions where alternative schemes suffer from low transmission, poor conversion efficiency or limited phase-matching bandwidths.

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